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Voltage-stabilised elastomers with increased relative permittivity and high electrical breakdown strength by means of phase separating binary copolymer blends of silicone elastomers

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1) Calculation of engineering stress and strain

The engineering stress (σ_E) was calculated from the force (*F*) and the cross-sectional area of the strip (*A*):

$$\sigma_E = \frac{F}{A} = \frac{F}{t \times w} = \frac{\tau \cdot d}{t \cdot w}$$

Equation 1

where *A* = film thickness (t) · constant width (w = 6 mm) and *F* = torque (τ) · drum diameter (*d* = 10.3 mm).

The engineering strain (ϵ_E) was calculated as a ratio of a stretched strain ($L - L_0$) to an initial strain (L_0) as:

$$\epsilon_E = \frac{L - L_0}{L_0}$$

Equation 2

where a final strain after stretching (*L*) was determined from Hencky strain (ϵ_H) as follows:

$$\epsilon_H = ln\frac{L}{L_0}$$

Equation 3

$$L = L_0 e^{\epsilon_H} = L_0 e^{(r_H . t_s)}$$

Equation 4

where ϵ_H is a product of Hencky rate (r_H = 1×10^{-3} rotation/s) and step time (t_s).

By putting equation (4) in (2), the final expression of engineering strain (ϵ_E) was obtained as below:

$$\epsilon_E = e^{\epsilon_H} - 1$$

Equation 5

Young's moduli were determined from slopes in the linear regime of stress-strain plots at 5 % strain.

2) NMR spectra of synthesised copolymers

The NMR spectra for synthesised PDMS-PPMS and PDMS-PEG copolymers are shown in Figures S1– S5.

a) PDMS-PPMS copolymer (**80DMS_2PMS**, $C_{C_6H_6}$ = 8.4 · 10^{-4} mol g^{-1})

^{1}H-NMR ($CDCl_3$, 300 MHz): δ -0.02 - δ 0.6 (m, 6 H's, -SiO(C**H$_3$**)$_2$-), δ 4.70 (m, 1 H, -Si**H**-), δ 7.10 - δ 7.60 (m, 5 H's, -SiC$_6$**H$_5$**-).

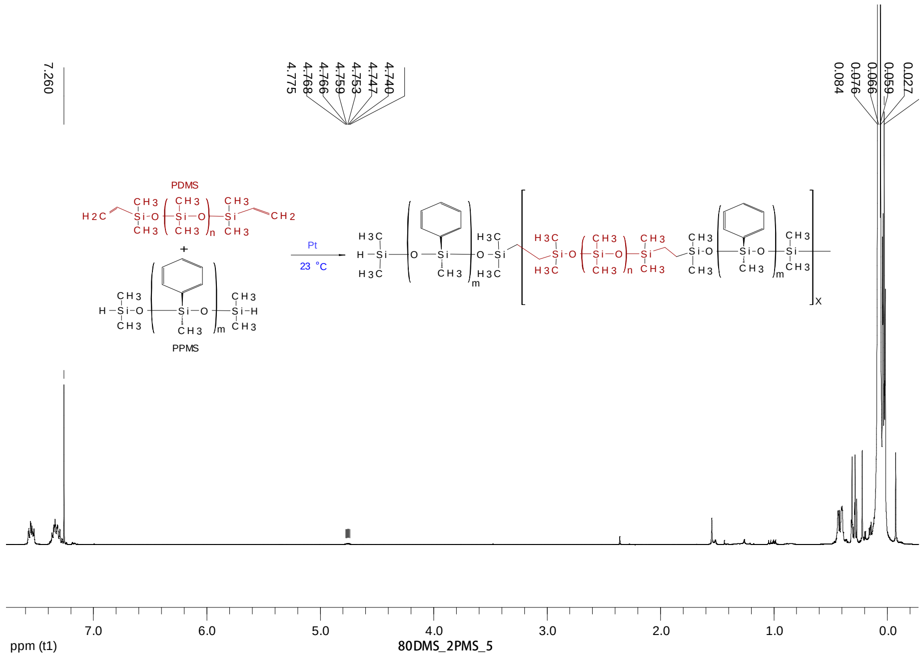


Figure S1 The NMR for 80DMS_2PMS.

b) PDMS-PEG copolymer (**PDMS81-PEG**)

^{1}H-NMR ($CDCl_3$, 300 MHz): δ 0.05 - δ 0.09 (m, 6 H's, -Si(C**H$_3$**)$_2$O-), δ 3.50 - δ 3.70 (m, 4 H's, -C$_2$**H$_4$**O-), δ 0.98 - δ 1.03 (t, 2 H's, -SiC**H$_2$**-), δ 3.53 - δ 3.57 (m, 2 H's, -CC**H$_2$**O-).

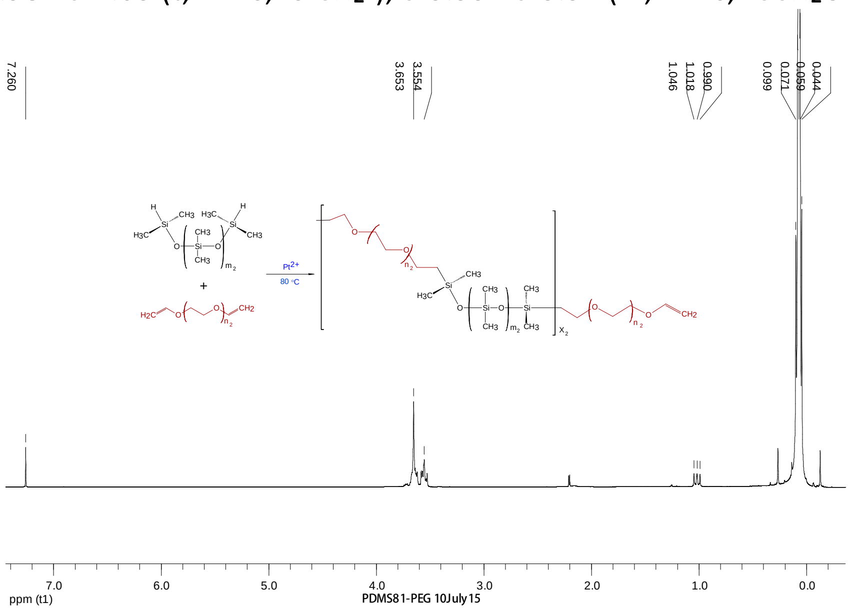


Figure S2 The NMR for PDMS81-PEG.

c) PDMS-PEG copolymer (**PDMS14-PEG**)

^{1}H-NMR ($CDCl_3$, 300 MHz): δ 0.05 - δ 0.09 (m, 6 H's, -Si(C**H_3**)**$_2$**O-), δ 3.50 - δ 3.70 (m, 4 H's, -C_2**H_4**O-), δ 0.98 - δ 1.03 (t, 2 H's, -SiC**H_2**-), δ 3.53 - δ 3.57 (m, 2 H's, -CC**H_2**O-).

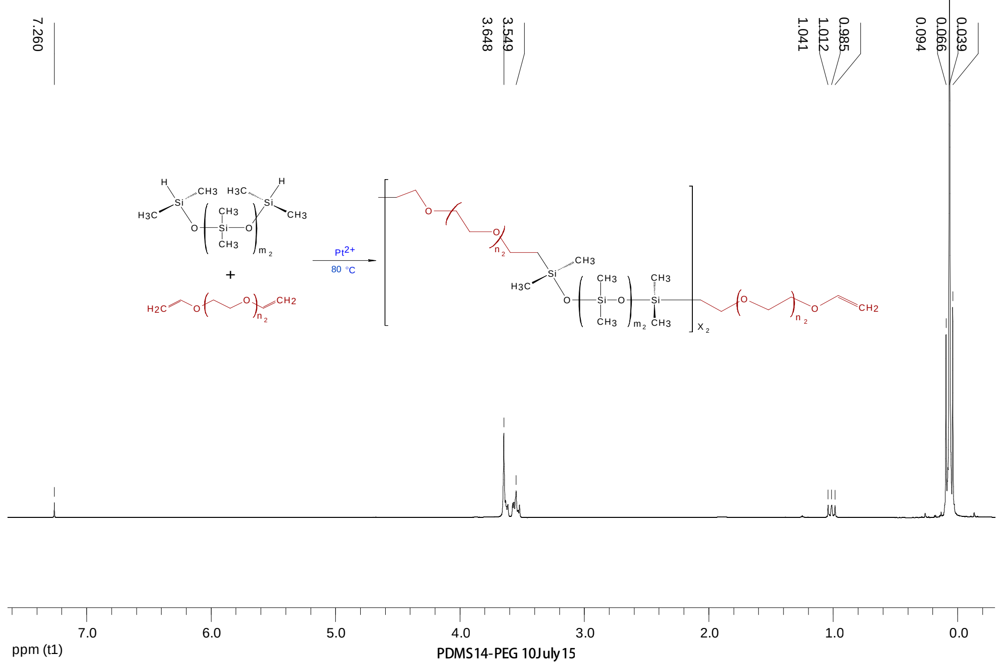


Figure S3 The NMR for PDMS14-PEG.

d) PDMS-PEG copolymer (**PDMS7-PEG**)

^{1}H-NMR ($CDCl_3$, 300 MHz): δ 0.05 - δ 0.09 (m, 6 H's, -Si(C**H_3**)**$_2$**O-), δ 3.50 - δ 3.70 (m, 4 H's, -C_2**H_4**O-), δ 0.98 - δ 1.03 (t, 2 H's, -SiC**H_2**-), δ 3.53 - δ 3.57 (m, 2 H's, -CC**H_2**O-).

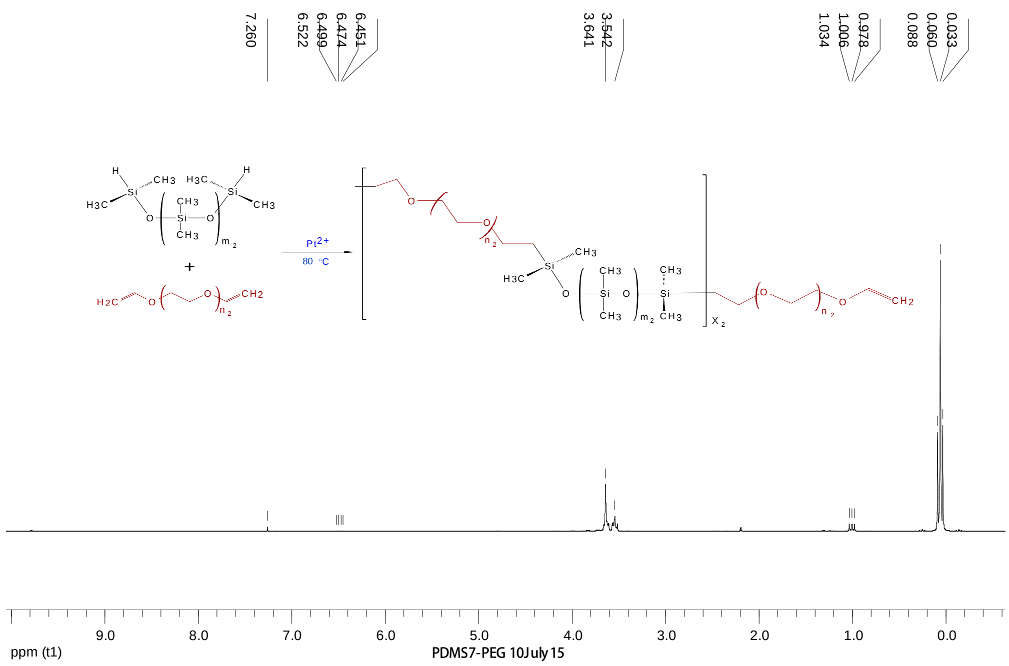


Figure S4 The NMR for PDMS7-PEG.

e) PDMS-PEG copolymer (**PDMS3-PEG**)

^{1}H-NMR ($CDCl_3$, 300 MHz): δ 0.05 - δ 0.09 (m, 6 H's, -Si(C**H$_3$**)$_2$O-), δ 3.50 - δ 3.70 (m, 4 H's, -C_2**H$_4$**O-), δ 0.98 - δ 1.03 (t, 2 H's, -SiC**H$_2$**-), δ 3.53 - δ 3.57 (m, 2 H's, -CC**H$_2$**O-).

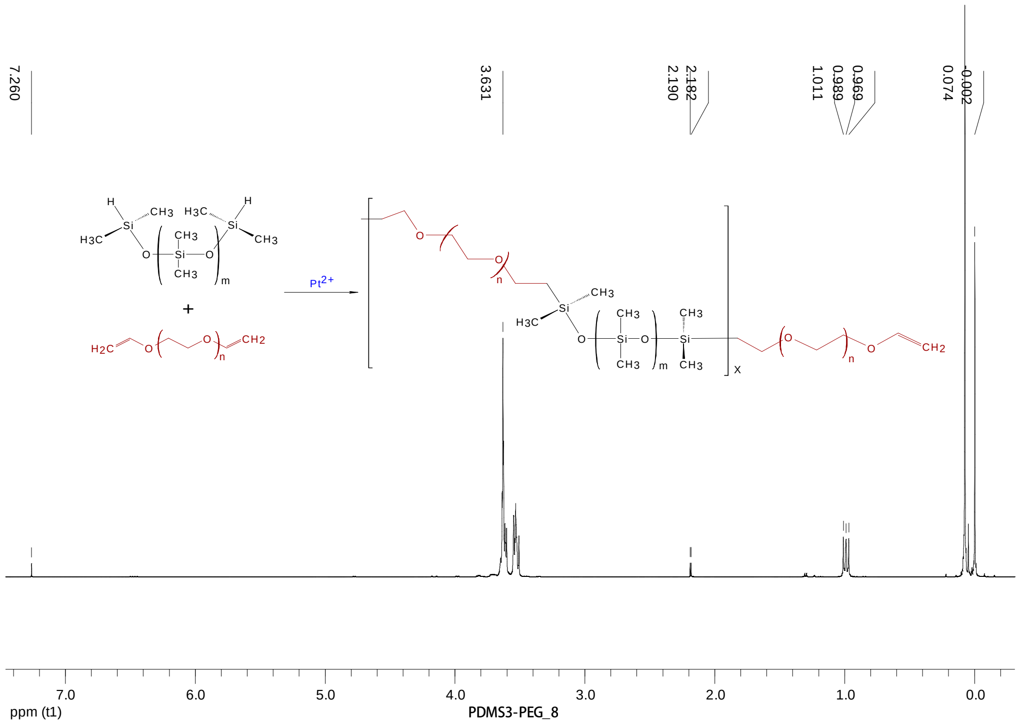
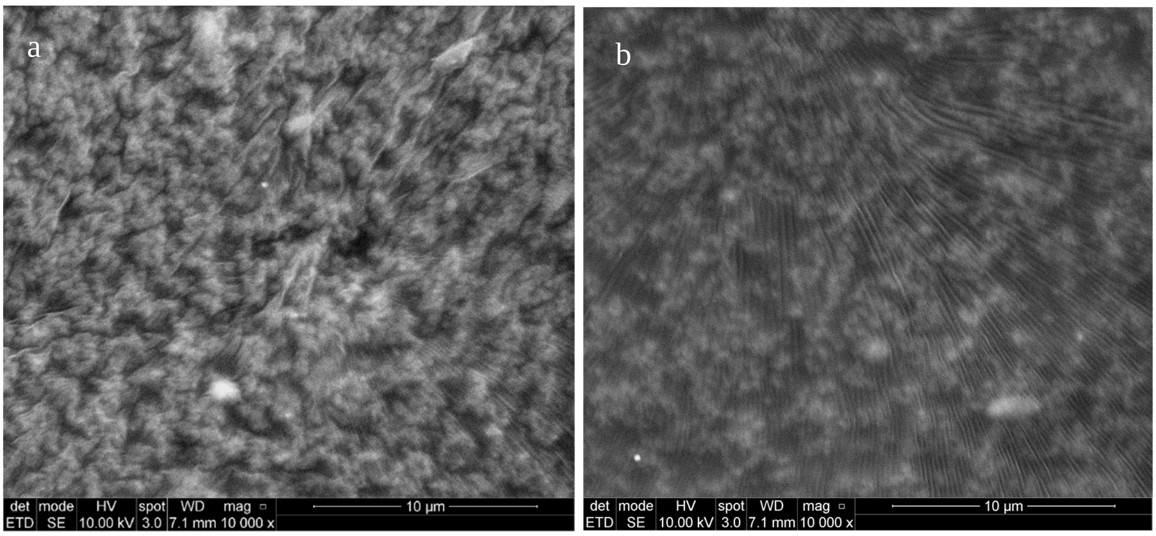


Figure S5 The NMR for PDMS3-PEG.

3) SEM images



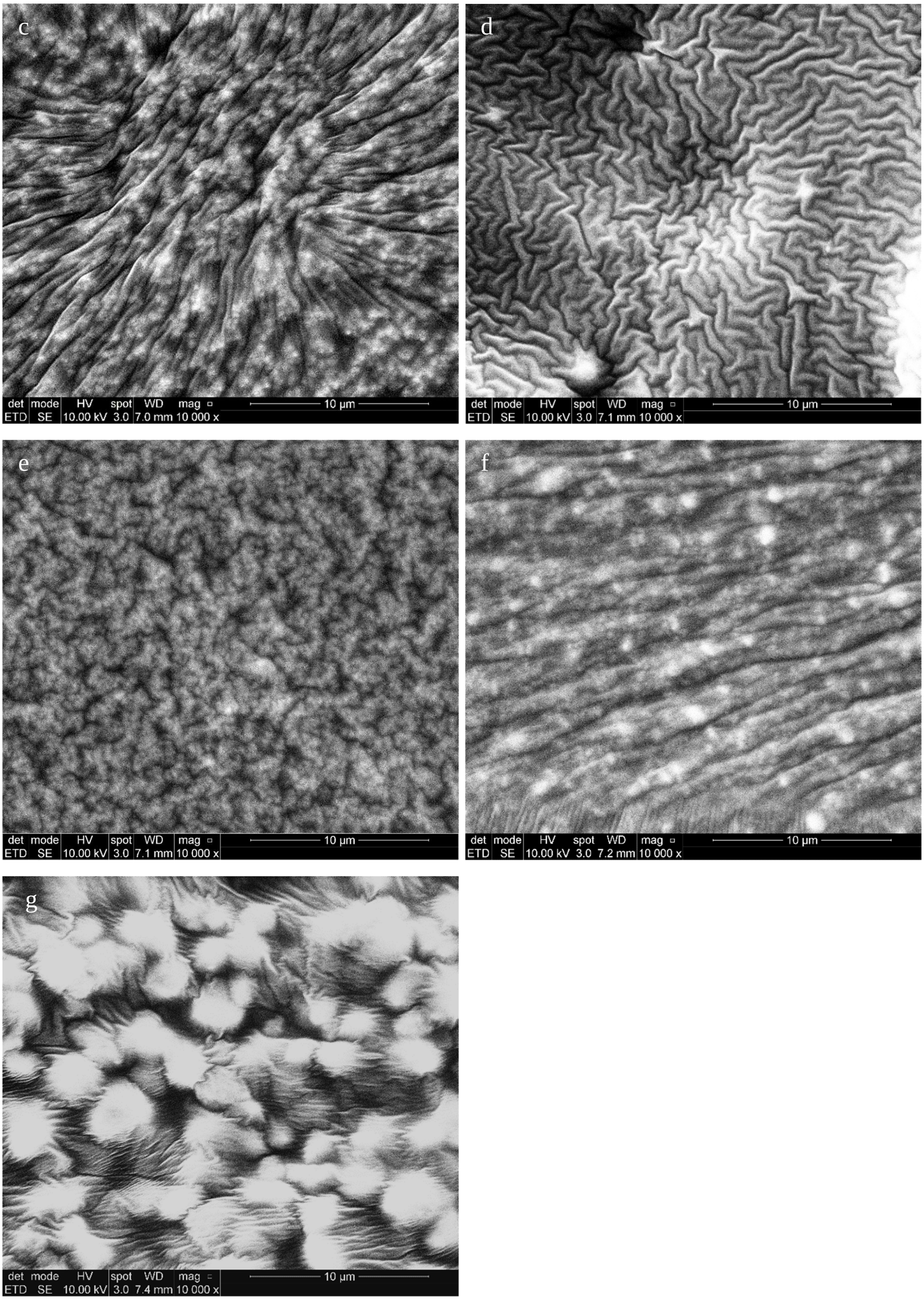


Figure S6 SEM images cross-linked BCBs with: **a**) 10 phr PDMS81-PEG, **b**) 20 phr PDMS81-PEG, **c**) 10 phr PDMS14-PEG, **d**) 20 phr PDMS14-PEG, **e**) 10 phr PDMS7-PEG, **f**) 10 phr PDMS3-PEG, and **g**) 20 phr PDMS3-PEG.